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Development of a Heat Conduction Model and Investigation on Thermal Conductivity Enhancement of AlN/Epoxy Composites

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Abstract

This work explores on the possibility of enhancement of heat conduction capability of a typical particulate filled polymer composite. A theoretical model for one dimensional heat conduction through such a composite has been developed using the law of minimal thermal resistance and equal law of the specific equivalent thermal conductivity. Based on this model, a mathematical correlation between the effective thermal conductivity of the composite and the filler content is proposed. The proposed model is then validated through experimentation conducted in controlled laboratory conditions. Different samples of aluminum nitride filled epoxy composites are prepared by simple hand layup technique. The content of the conductive filler (AlN) in the composites is varied in the range 0% to 15% by volume. Thermal conductivities of these composite samples are measured according to the ASTM standard E-1530 by using the Unitherm™ Model 2022 tester, which operates on the double guarded heat flow principle. Gradual increase in thermal conductivity has been observed, with increase in conductive filler percentage in composite samples. Values obtained using the proposed mathematical correlation are then compared with the measured experimental results and also with values estimated with other established correlations such as Rule-of-Mixture (ROM), Maxwell's model, Bruggeman model and Nielson-Lewis model. This comparison reveals that while none of the above models are correctly predicting the effective thermal conductivity of the composites, the results obtained from the proposed model by incorporating a correction factor are in good agreement with the experimental data.

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Keywords: Polymer Composites; Aluminium Nitride; Thermal Conductivity; Theoretical Model

Nomenclature

H	:	Side length of the cube element
r	:	Radius of spherical filler
dT	:	temperature difference between two side of the element
k_p, k_f	:	thermal conductivity of polymer matrix and filler material
k_{eff}	:	effective thermal conductivity of the composite material
S_p, S_f	:	cross sectional area of the matrix material and filler material in the element
S	:	cross sectional area of the composite material for an element
Q_p, Q_f	:	heat flow through the cross sectional area of matrix and filler in the element
Q	:	heat flow through the cross sectional area of an element of composite material
R	:	total heat resistance of the element
V_p, V_f	:	Volume of matrix material and filler material in the element
V	:	Volume of composite material for an element
Φ_f	:	Volume fraction of the filler

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1. INTRODUCTION

Polymers and polymer matrix composites (PMCs) are the most common materials currently being utilized by several industries as they show remarkable properties when it comes to corrosion resistance, durability, low density and low fabrication cost etc. But owing to their low thermal conductivity, they often become the secondary choice for certain applications. For example, the requirement of electronic industry for using light weight materials with high heat conductivity is largely fulfilled by expensive metallic materials or metal matrix composites. As the demands in denser and faster circuits intensify, the heat dissipation in microelectronic packaging becomes increasingly important. And traditionally, thermal problem in encapsulated devices has been addressed by the use of high cost embedded heat sinks, which are often susceptible to thermal cracking and of limited utility in thinner packages [1]. Under this circumstance, polymers filled with thermally conductive fillers are emerging as a cost effective ways to cope with such thermal management issues [2]. PMCs with improved conductivity would certainly serve as alternate options. Reinforcement of filler in polymers is thus becoming a common practice in electronic industries. As already mentioned, with the rapid development of the electronic information industry, better properties are required for substrate and packaging material, such as high thermal conductivity, low coefficient of thermal expansion and low dielectric constant. Polymer composite materials have been found extremely useful for such heat dissipation applications [3, 4]. By reinforcing metal particulates with high thermal conductivity into the polymers, composites with intermediate conducting property [5, 6] can be obtained. The above interest arises from the fact that while the thermal and electrical properties of such composites approach towards metal, the mechanical properties and processing method are those of polymers. To effectively solve the thermal dissipation problem, addition of different fillers in polymeric matrices has been investigated. This includes oxides such as Al_2O_3, ZnO [7], carbide such as SiC [8], nitrides like AlN and BN [9-10] and carbon materials such as carbon nanotubes [11]. Among these fillers, AlN has been considered an ideal candidate due to its very high thermal conductivity, no toxicity, stable crystal structure and relatively low cost. In addition to thermal conductivity improved by AlN filler, other properties such as CTE, tensile strength, and dielectric constant of polymer can also be modified by adding AlN because of its low CTE, high elastic modulus and relatively low dielectric constant. Thus, AlN seems to be an excellent filler to improve the properties of polymers for application in substrate and packing materials.

In view of this, the present work has been undertaken to study the effect of adding micro-sized AlN particles on the thermal conductivity of epoxy resin and to develop a heat conduction model using the law of minimal thermal resistance and equal law of the specific equivalent thermal conductivity. Based on this model, a correlation between the effective thermal conductivity of the composite and the filler content is proposed. The proposed model is then validated through experimentation conducted in controlled laboratory conditions.

2. MODELS FOR EFFECTIVE THERMAL CONDUCTIVITY FOR COMPOSITES

To predict the effective thermal conductivity of composite materials there are several theoretical and empirical models have been proposed. There are many analytical model studies on thermal conductivity of filled polymer composites. The two basic equation of thermal conduction used in composites are Rule of mixture derived on the basis of series conduction and parallel conduction respectively, Maxwell [12] assumes a random dispersion of small sphere within a continues matrix to calculate the effective thermal conductivity which hold good for low filler concentrations. Bruggeman [13] derived an equation of thermal conductivity versus the solids loading for spherical fillers in a dilute suspension. Lewis and Nielsen [14] modified the effect of the shape and the orientation of the particles for a two phase system. A few comprehensive review articles too have discussed the applicability of many of these models [15-16]. For a two-component composite, the simplest alternatives would be with the materials arranged in either series or parallel with respect to heat flow.

For the parallel conduction model:

$$k_{eff} = \left(1-\phi_f\right)k_p + \phi_f k_f \tag{1}$$

For series conduction model

$$\frac{1}{k_{eff}} = \frac{\left(1-\phi_f\right)}{k_p} + \frac{\phi_f}{k_f} \tag{2}$$

The correlations presented by Eqs. (1) and (2) are derived on the basis of the Rules-of-mixture. Maxwell [12] obtained an exact solution for the conductivity of randomly distributed and non-interacting homogeneous spheres in a homogeneous medium

$$\frac{k_{eff}}{k_p} = \left(\frac{k_f + 2k_p + 2\phi_f\left(k_f - k_p\right)}{k_f + 2k_p - \phi\left(k_f - k_p\right)}\right) \tag{3}$$

Thermal conductivities for low filler concentrations are predicted very well using this model; but when there is an increase in filler concentrations, conductive chains is started to form because the particle starts to come in contact with each other, this conducting chain is in the direction of heat flow, that is why k_{eff} are underestimated for the above model. Some other models which can be used for predicting the effective thermal conductivity of polymer composite system are Bruggeman model

$$1-\phi_f = \frac{k_{eff}-k_f}{k_p-k_f}\left(\frac{k_p}{k_{eff}}\right)^{\frac{1}{3}} \tag{4}$$

and Nielson-Lewis model

$$k_{eff} = k_p\left(\frac{1+AB\phi}{1-B\phi\psi}\right)$$

Where A and B are the constants and are having a specific values for a particular shape and size of filler particles.

3. DEVELOPMENT OF A NEW THEORETICAL MODEL

Fig. 1 shows the 3-D view of particulate filled composite cube and a single element is taken out from it for further study the heat transfer behavior as shown in Fig. 2 consisting of a small cube with a single particle is in the center of it.
The theoretical analysis of heat transfer in composite material is based on the following assumption:

(a) Locally both the matrix and filler are homogeneous and isotropic.
(b) The thermal contact resistance between the filler and the matrix is negligible and the lamina is free from voids.
(c) The temperature distribution along the direction of heat flow is linear.

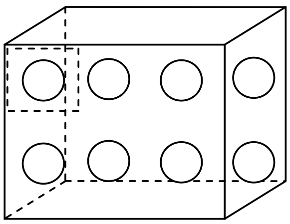

Fig.1 3-Dimensional view of particulate filled composite cube

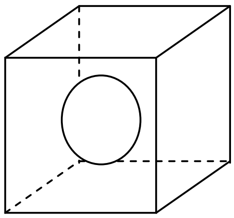

Fig.2 3-Dimensional view of element under study

On the basis of law of minimal thermal resistance and equal law of the specific equivalent thermal conductivity, when only one mode of heat transfer is considered i.e. heat conduction, and specific equivalent thermal resistance of single element of the composite is considered equal to the total thermal resistance of the composite, then the equivalent thermal conductivity of that single element is considered equal to the total thermal conductivity of the composite, and while considering that it is not necessary to consider the size of the element. Fig. 3 shows the front view of element under study having side length *H* and a single spherical particle of radius *2r* at the centre, the combination of such small cube element is overall composite, the direction of flow of heat is considered from top to bottom.

The different thermal property of a body depends on the path followed by the heat while transferring in the materials. A series model of heat conduction through the unit cell of particulate filled polymer composite is considered as shown in Fig. 4. The element is divided into three parts, Part I and part III represents the neat polymer while part II represent the combination of polymer matrix and particle. k_1, k_2 and k_3 are the mean conductivity coefficient of respective parts. The thickness of part I and part III are h_1 and h_3 respectively, for simplicity both the above thickness is considered to be equal and $h_1 = h_3 = H-2R$. Part II having a thickness $h_2 = 2R$. To determine the effective thermal conductivity of the whole element, the law of minimum thermal resistance is required to combine the heat resistances of these three parts to get the heat resistance of the complete element and the equal law of specific thermal conductivity is applied to predict the effective thermal conductivity of the complete element. As already assume the linear distribution of temperature, thermal conductivity of each section can be calculated as:

For part I and III:
Since no filler particle is there in that region, so thermal conductivity of that region will be same as of polymer matrix i.e.

$$k_1 = k_3 = \int_{h1} k_p \frac{dy}{h_1} = k_p \tag{5}$$

For part II:
Taking a thin piece with thickness dy, applying Fourier's law of heat conduction, k_2 is

$$k_2 = \frac{Q_p + Q_f}{\left({dT}/{dy} \right) S}$$

$$k_2 = \int_{h_2} \frac{\left(k_p S_p / S + k_f S_f / S \right) dy}{h_2} = \frac{1}{h_2 S}\left(k_p V_p + S_f V_f \right) \tag{6}$$

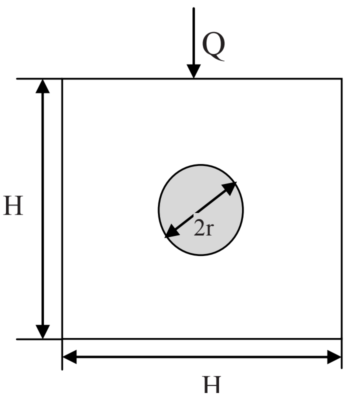

Fig.3 Physical model of heat transfer

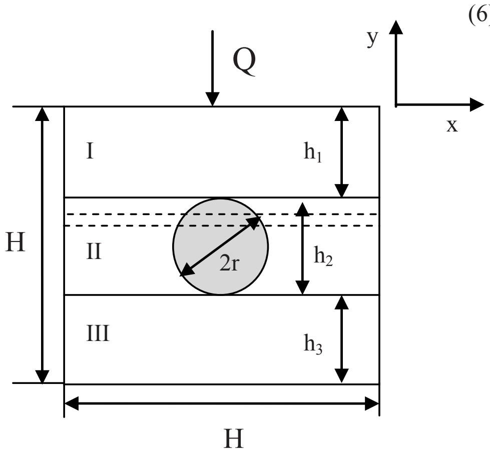

Fig.4 Series model of heat transfer

Similarly thermal resistance of the three parts is

$$R_1 = R_3 = \frac{h_1}{k_p S} \tag{7}$$

$$R_2 = \frac{h_2}{\frac{1}{h_2 S}\left(k_p V_p + k_f V_f \right) S} = \frac{h_2^2}{k_p V_p + k_f V_f} \tag{9}$$

As the series model is considered for heat transfer in the element, the k_{eff} of composites is given by

$$k_{eff} = \frac{H}{RS} = \frac{H}{\left(R_1 + R_2 + R_3 \right) S}$$

The above equation is written on the basis of micro sized spherical particles being uniformly distributed in the polymer matrix, but practically uniform distribution of particles are not possible, also it is assumed while writing the equation that heat flows in one direction only but in reality while measuring the thermal conductivity it is impossible to restrict the heat flow direction to one. Because of the above assumptions a correction factor ξ is incorporated which would count for all these discrepancies.

$$k_{eff} = \xi \frac{H}{\left(\frac{h_1}{k_p S} + \frac{h_2^2}{k_p V_p + k_f V_f} + \frac{h_1}{k_p S} \right) S} \tag{10}$$

Substituting equations (5) to (9) into equation (10), the k_{eff} of the polymer composite material is given by:

$$k_{eff} = \xi \frac{1}{\frac{1}{k_p} - \frac{1}{k_p}\left(\frac{6\phi_f}{\pi} \right)^{\frac{1}{3}} + \frac{4}{\left(k_p \left(\frac{4\pi}{3\phi_f} \right)^{\frac{2}{3}} + \left(\frac{2\phi_f}{9\pi} \right)^{\frac{1}{3}} 2\pi \left(k_f - k_p \right) \right)}} \tag{11}$$

The correlation given in equation (11) can be used to estimate k_{eff}, by suitably tuning the value of ξ .

3. EXPERIMENTAL DETAILS

Epoxy LY 556 resin the corresponding hardener (HY 951) is mixed in a ratio of 10:1 by weight as recommended. The epoxy resin and the hardener used for the present work are supplied by Ciba Geigy India Limited. Epoxy is chosen primarily because it happens to the most commonly used polymer and because of its low density (1.1 gm/cc) and insulating nature (low thermal conductivity, about 0.363 W/m-K). Micro-sized AlN powders obtained from synthesis by carbothermal reduction of α-Al_2O_3 (99.5 % purity) in ammonia using extended arc open plasma reactor is used as the filler material in the present investigation. AlN melts at 2800 °C in inert atmospheres, shows its stability at high temperatures. In air, AlN surface oxidation occurs above 700°C and in vacuum it decomposes at much higher temperature of around 1800. Up to 980°C it is stable in hydrogen and carbon dioxide atmospheres as well. AlN is chosen because of its conducting nature (high thermal conductivity, about 160 W/m-K).The room temperature curing epoxy resin (LY 556) and the corresponding hardener (HY951) are mixed in a ratio of 10:1 by weight as per recommendation. Aluminium nitride particles with average size 90-100μm are reinforced in epoxy resin having density 1.1 gm/cc to prepare the composite specimens. AlN used in this work possess density of 3.3 gm/cc. The composite specimens are cast by conventional hand-lay-up technique. Composites of seven different compositions with 0vol%, 2.5vol%, 5vol%, 7.5vol%, 10vol%, 12.5vol%, and 15vol% respectively are prepared. Unitherm™ Model 2022 is used to measure thermal conductivity of a variety of materials. The tests are in accordance with **ASTM E-1530** standard.

4. RESULTS AND DISCUSSION

Table 1 gives effective thermal conductivity of the AlN- epoxy composite for volume fraction ranging from 0% to 15%. For a given AlN of 100 micron particle size, the effective thermal conductivity of the composite material increases as the volume fraction is increasing. The table shows the comparison of effective thermal conductivity of AlN based epoxy composite calculated from various established results with the measured experimental values and the new theoretical model. These results are graphically shown in Fig. 5. It shows the comparison between the various established theoretical model, experimental results and the proposed model. It is evident from this figure that there is appreciable increase in thermal conductivity as the concentration of AlN particle is increasing. Though the values obtained from the established theoretical model are quite low as compared to the measured values, but it can be seen that the variation of the theoretical estimation by proposed model of the effective thermal conductivity is similar to the experimental measured values.

Table 1 Effective thermal conductivity values for composites obtained from different methods

Sample	Filler Content	Effective thermal conductivity of composites K_{eff} (W/m-K)					
		Rule of mixture	Maxwell's equation	Bruggeman's model	Lewis and Nielsen's equation	Proposed model	Experimental value
1	2.5	0.372	0.390	0.392	0.386	0.414	0.418
2	5	0.382	0.419	0.425	0.412	0.485	0.492
3	7.5	0.392	0.450	0.459	0.440	0.552	0.561
4	10	0.403	0.483	0.498	0.472	0.621	0.611
5	12.5	0.414	0.517	0.542	0.507	0.692	0.661
6	15	0.426	0.553	0.592	0.547	0.770	0.712

It can be seen in Fig. 6 that for proposed model, as the volume fraction of the filler is increasing above 40 %, there is a rapid increase in the value of thermal conductivity. The reason for sudden increase in K_{eff} value after certain percentage is that, if we consider a single element, volume fraction is increasing means radius of the spherical ball is increasing, as its radius increasing it try to fill the cube element reducing the matrix material, results in the increase in particle interaction, and after particular value i.e. H=2r spherical ball touches the cube boundary.

It can be observed that the value of effective thermal conductivity turns to negative value as the volume fraction increased beyond 55%. The reason for that is, as the volume fraction increases, for a single element, the radius of the spherical particle has to increase, after a certain value H becomes less than diameter of the spherical particle and particle comes out of the cube, but if we combine such cube there will be interference between the particle and they overlap each other which is not possible practically. This is the reason the proposed model is giving absurd value. So the proposed model holds good for volume fraction less than 55%. Liang [17] also describes such relationship and found that the model holds well when the filler volume fraction is less than 60%.

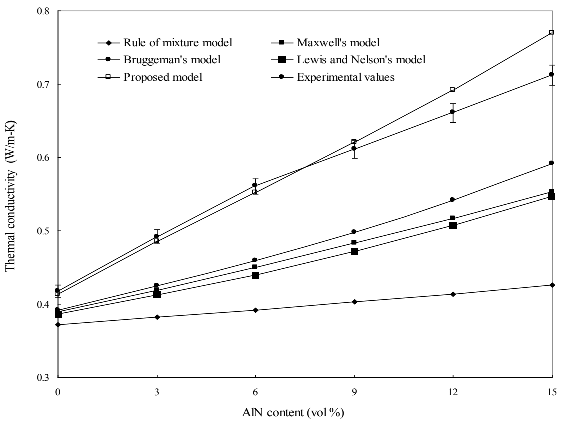

Fig. 5 Thermal conductivity of AlN/Epoxy: Rule of mixture, Maxwell's model, Bruggman model, Nielson-Lewis model, proposed model and Experimental values

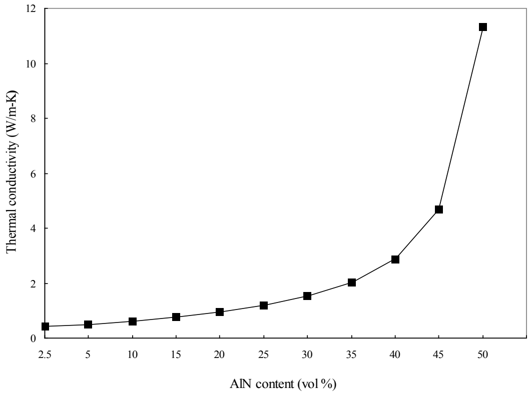

Fig. 6 Variation of effective thermal conductivity with volume fraction for proposed theoretical model

5. CONCLUSIONS

A theoretical model of one dimensional heat conduction has been proposed, which successfully estimates the effective thermal conductivity of typical particulate filled polymer composite systems with filler volume fraction up to 55%. The values obtained from this model are found to be in good agreement with the experimental results for a set of aluminum nitride filled epoxy composites. It is suggested that this correlation can be further used for assessment of the effective conductivity of any other similar particulate-polymer composite systems by suitably tuning the value of correction factor. This work shows that as the volume fraction of the AlN in the composite is increased, the thermal conductivity of the composite also increases and the rate of increase of thermal conductivity is rapid for high volume fraction i.e. above 35% as compared with low volume fraction. With light weight and improved heat conduction capability, these AlN filled epoxy composite can be used for applications such as electronic packages, communication device, thermal grease, thermal interface material and electric cable insulation.

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